RADIONUCLIDE MIGRATION IN PLUTONIC ROCKS: IMPLICATIONS FOR HIGH-LEVEL NUCLEAR WASTE DISPOSAL.

Vanessa Ann Guthrie

B.Sc. (Hons.) Dip. Nat. Res.

submitted in fulfillment of the requirements for the degree of

Doctor of Philosophy.

University of Tasmania

Hobart, Tasmania.

February 1991.

This thesis contains no material which has been accepted for the award of any other higher degree or graduate diploma in any tertiary institution, and to the best of my knowledge and belief, no copy or paraphrase of material previously written or published by another person, except where due reference is provided.

Vanessa A Guthrie

<u>*Vanessa Ann Guthrie.*</u>

February 1991.

It is our responsibility to ensure that we leave for our children

the environment which we hold in trust not as a legacy

but as a protected and well managed resource.

Let us aim to meet the challenge.

Abstract

Geochemical studies of deeply buried intrusive rocks provide the foundation for evaluating the suitability of crystalline rocks as repositories for solidified High-Level Nuclear Waste (HLW). This geochemical study examines the migration of natural and introduced radionuclides during interaction between groundwater and plutonic rock, to provide an understanding of the processes which may operate in the potential repository environment.

Rock cores from the Coles Bay Granite (Tasmania), Kambalda Granodiorite (Western Australia) and the Roxby Downs Granite (South Australia) were selected for this study on the basis of the variation in mineralogy, fracture density and degree of alteration. Firstly, the behaviour of U- and Th-decay series radionuclides in each rock was investigated as a natural analogue for some HLW elements to identify the nuclide migration pathways and significant sites of rock/radionuclide interaction. Secondly, Synroc doped with actinides and fission products was used as a source of radionuclides to evaluate the interactions between waste elements and intrusive rocks in a simulated water-saturated repository environment. This integrated approach has identified the mechanisms controlling radionuclide release, migration and retention.

In the natural analogue studies, the application of fission-track micromapping has determined that primary uranium is distributed in the three intrusives as background U in the major rock-forming minerals, and as resistate U in the primary accessory phases. Two modes of redistribution of this uranium are evident; as secondary U in the secondary minerals formed during alteration, and as fracture U associated with the fracture-infilling minerals. The mechanisms for uranium retention are dominated by adsorption and ion exchange. The study of uranium-series disequilibrium has determined that significant radionuclide mobilisation has occurred in the recent past (<1.2 Ma) as a result of groundwater interaction. Disequilibrium between ^{230}Th, ^{234}U and ^{238}U in the three intrusives indicates that fractures form the

dominant nuclide migration pathways and the most significant sites of rock/radionuclide interaction are the secondary and fracture-infilling minerals.

Leach testing of Synroc with the three intrusives was carried out to determine the mechanisms and processes which occur during Synroc/water/granitic host rock interaction. Significant geochemical and mineralogical changes were observed in all three intrusives during leach testing, including loss of crystal structure and formation of surface reaction products. These changes are reflected in a change in the leach solution conditions and may also affect the distribution of radionuclides during leach testing. The presence of the intrusives significantly inhibited the total release of the actinides (Np, Pu and Cm) and the less soluble fission products (Zr, Ce, Nb and Ru) from Synroc, as a result of the change in solution chemistry and nuclide solubility induced by the presence of the granites. Substantial preferential uptake of all radionuclides by specific secondary and fracture infilling minerals (such as sericite, hematite, Fe- and Ti-oxides/hydroxides) in intrusives was also observed, which was controlled by rapid ion exchange, redox reactions, sorption and surface deposition of colloids and pseudocolloids.

These results imply that fractures will form the main pathways for radionuclide mobilisation in the HLW disposal environment, and that the most significant sites for rock/radionuclide interaction will be the secondary and fracture-infilling minerals. Sorptive processes will dominate radionuclide retention and therefore retard migration of these elements away from the waste package into the surrounding near-field geological environment. The geochemical evolution which occurs during rock/water interaction may affect radionuclide release, migration and retention through changes in solution characteristics and sorptive capacity. These qualitative experimental observations of the chemically complex interactions which occur in the predicted repository environment may be used for quantitative predictive modelling in repository assessment.

Acknowledgements

To see the completion of this thesis is the fulfillment of a dream. Many people have assisted me in the progress of this work, have believed in my capabilities at times when I did not believe in them myself, and have consistently and unstintingly given of themselves throughout the duration of the work. I would like to thank all of these people, and in particular;

- *at the Australian Institute of Nuclear Science and Engineering;*

Roger Gammon, and his predecessor, **Bill Palmer,** for their enthusiasm and encouragement of my work,

- *at the Australian Nuclear Science and Technology Organisation;*

Des Levins, for his enthusiasm, guidance and insight in establishing the project, and encouragement and support of me to persevere

Kaye Hart, for her scientific ability, understanding and patience in teaching me in the face of all confusion, her constant encouragement to me to continue and, not least of all, her friendship and support

Kath Smith, for her time and effort spent in patient explanation, her guidance in TEM techniques and her friendship

Brian Seatonberry, for his ability to make sense of my garbled notes and assistance in producing consistent results

Adam Jostsons, for his interest in and encouragement of the project from its inception.

Others who have offered their assistance without question include Sue Brown, John Warmeant, Brett Robinson, Roy Warren and Dim Roman. Many thanks.

- *at the University of Tasmania;*

Joe Stolz, for his personal and professional support, enthusiasm and encouragement (sometimes at his own expense), his scientific insight and quick understanding of confounding problems, his time and effort spent reading through earlier versions of each chapter, and for his ability to ask curly questions when you least expect them

Russell Sweeney and Graeme Wheller, for their efforts spent in reading some early drafts of both papers and chapters

Rick Varne, for his time and effort in seeing my thesis through the mill

Dave Green, for his continued interest and encouragement of this work and my professional career

Garry, Steve, Michael and June, for their friendship and constancy, and the other post-graduate students and post-doctoral fellows of the Department of Geology for their interest and support. You know who you are — thanks.

Access to core samples was arranged by staff of the *Tasmanian Department of Mines, and Western Mining Corporation — Kambalda Nickel Operations and Olympic Dam Project.* In particular, I wish to thank **Ken Cross,** for his time and effort in arranging access to samples at Olympic Dam, and reviewing earlier drafts of papers and chapters.

This work was undertaken while I was in receipt of an AINSE studentship, without which the work would never have been completed. Western Mining Corporation and Australian Nuclear Science and Technology Organisation, Materials Division and Chemical Engineering Section also provided financial and logistic support for which I am very grateful. Papers arising from this work have been published with the kind permission of AINSE, ANSTO and WMC — Olympic Dam Project and Kambalda Nickel Operations.

Finally, I would like to express my appreciation to all of my friends, including my parents, both scientists and non-scientists alike, for their continued support throughout the duration of this work. Thanks.

Table of Contents

SECTION IV: *IMPLICATIONS AND CONCLUSIONS*

Chapter 9: *Implications for HLW Disposal*

Chapter 10: *Conclusions*

SECTION V: *REFERENCES AND APPENDICES*